

VIBRONIC MECHANISM OF THE ISOTOPE AND PRESSURE EFFECTS IN CUPRATE

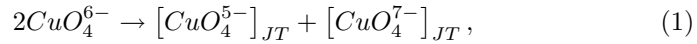
A. S. MOSKVIN, Y. D. PANOV

*Department of Theoretical Physics, Ural State University,
Lenin av.51, 620083, Ekaterinburg, Russia
E-mail: alexandr.moskvin@usu.ru*

The doped cuprates are considered to be a system of local spinless bosons moving in a lattice of the hole pseudo-Jahn-Teller centers CuO_4^{5-} . A detailed qualitative and quantitative description of the vibronic mechanism determining both the isotope substitution and the external pressure effect on T_c is given. This mechanism can properly interpret the principal peculiarities of isotopic and baric effects in cuprates except the region of the well developed percolation phenomena

Intuitive ideas concerning an especial role played by the Jahn-Teller (JT) ions (centers, polarons) have been used as a starting point of the pioneer investigations by K.A. Müller and J.G. Bednorz accomplished in 1986 by the outstanding discovery of the high- T_c superconductivity. One of the most realistic JT-effect based scenarios for doped cuprates like $La_{2-x}Sr_xCuO_4$ was suggested and elaborated earlier¹. The most exciting features of the hole centers CuO_4^{5-} are associated with an unusually complicated ground state resulting from the electronic quasi-degeneracy accompanied by the (pseudo)-Jahn-Teller (PJT) effect. An additional hole doped to the basic CuO_4^{6-} cluster with the b_{1g} hole can occupy both the same hybrid $Cu3d - O2p$ orbital state resulting in a Zhang-Rice singlet $^1A_{1g}$ and the purely oxygen e_u molecular orbital resulting in the singlet or triplet $^1,^3E_u$ terms with the close energies. If the vibronic coupling is strong enough, the adiabatic potential (AP) of the hole $[CuO_4^{5-}]_{PJT}$ PJT center has the four equivalent minima, which correspond to the hybrid distortions of the $b_{1g}e_u$ - or $b_{2g}e_u$ -type, so the vibronic ground state is fourfold degenerated, if to neglect a tunnel splitting. The hole PJT center with its high polarizability could be a center of an effective local pairing with a formation of a novel electron PJT center $[CuO_4^{7-}]_{PJT}$.

A novel superconducting phase in the doped cuprates appears to be a result of the disproportionation reaction



with formation of system of the local spinless bosons moving in a lattice of the hole PJT centers or the quantum lattice bose liquid. Bose-condensation temperature is proportional to the local boson transfer integral: $T_c = t_{BB}f(N_B)$,

where $f(N_B)$ is a smooth function of the boson concentration². The PJT nature of the polar (electron and hole) CuO_4 clusters provides a main vibronic mechanism both of the isotopic and baric effects on the T_c due to vibronic reduction of transfer integral³

$$t_{BB} = t_{BB}^{(0)} K_{eh} \quad (2)$$

with the vibronic reduction factor K_{eh} , or the Franck-Condon factor

$$K_{eh} = \langle \Psi_e | \Psi_h \rangle^2, \quad (3)$$

where Ψ_e and Ψ_h are the ground state vibronic functions in a definite AP minimum for electronic (e -) and hole (h -) center, respectively. These functions depend on different parameters of the polar center (such as elastic and vibronic constants, crystal field parameters), so that any impact on the lattice or electron subsystems can change the K_{eh} magnitude. Real perspectives for a high- T_c superconductivity should be connected with an extremely weak inter-mode coupling regime or with the so called *optimized systems*. A boson movement in optimized systems is not accompanied by a significant variation in the spin and the local structure modes or in other words the charge fluctuations do not result in the spin and structure fluctuations. Electron and hole PJT centers in optimized systems have identical vibronic states with the maximal value of the "vibronic reduction" factor $K_{eh} = 1$, so the vibronic contribution to the T_c variation (suppression or enhancement) for these systems due to the isotope substitution, the external pressure application etc., is entirely suppressed. In practice, the specific features of the optimized systems are the maximally high T_c 's, the minimal width of the superconducting transition, the minimal magnitude of the isotope effect, the minimal value of the baric coefficient dT_c/dp . Probably, the so called "optimally doped" $YBa_2Cu_3O_{6+x}$ oxide at $x \approx 0.93$ can be approximately considered as one of the real optimized systems. A detailed analysis of the PJT effect within the ($^1A_{1g}, ^{1,3}E_u$) manifold with account of the whole set of the active displacement modes of the a_{1g} , b_{1g} , b_{2g} , e_u symmetry allows to obtain an analytical form for the reduction factor K_{eh} . This can be reduced to universal form:

$$K_{eh} = D \exp(-g^2), \quad (4)$$

where D and g in a complicated manner depend on the parameters like bare energy separations within ($^1A_{1g}, ^{1,3}E_u$)-manifold, vibronic constants, the oxygen and copper atomic masses. Detailed analytical examination of general expressions for g and D with account of real multi-mode situation is very complicated. But a case of isotope substitution appears to be essentially simplified because

of only vibrational part of K_{eh} plays a role. For a simplest one-dimensional single-mode situation

$$K_{eh} = \frac{2\tau}{1 + \tau^2} \exp\left(-\frac{(\Delta Q)^2}{l_e^2 + l_h^2}\right), \quad (5)$$

where l_e and l_h are the effective oscillatory lengths of the e - and h -centers, respectively, $\tau = l_e/l_h$, $g^2 \sim (\Delta Q)^2$, with ΔQ being the distance separating the AP minima for the e - and h -centers. For two-dimensional oscillators centered for simplicity at the same point $g = 0$, thus $K_{eh} \equiv D$ and

$$K_{eh} = 4 \left[\tau + \frac{1}{\tau} + \alpha \cos^2 \Delta\phi + \beta \sin^2 \Delta\phi \right]^{-1},$$

$$\alpha = \left(\frac{\tau_e}{\tau_h} + \frac{\tau_h}{\tau_e} \right), \beta = \left(\tau_e \tau_h + \frac{1}{\tau_e \tau_h} \right), \quad (6)$$

where $\tau_i = l_i^{(1)}/l_i^{(2)}$ ($i = e, h$), $l_i^{(1,2)}$ - oscillatory lengths, which are the principal axes of the polarization ellipse defined by quadratic form for AP expansion near potential minima for e - or h -center, respectively; $\tau = S_e/S_h$, S_i is an area of ellipse; $\Delta\phi$ is an angle between principal axes of ellipses for e - or h -center.

So, in a multi-mode case the g contribution plays the same role, but D factor provides two contributions to vibronic reduction, due to variation in the oscillatory lengths ("*stretching*" mode) and due to variation in relative orientation of the polarization ellipses ("*tilting*" mode) for the e - or h -center, respectively.

For isotope effect (IE) this leads to the following results. In the simplest one-mode case l_e and l_h have an identical dependences on mass: $l_{e,h} \sim m^{-\frac{1}{4}}$, and thus D factor doesn't depend on atomic masses. Another factor $g^2 \sim \sqrt{m}(\Delta Q)^2$, so that $\alpha \geq 0$ and IE will be determined mainly by the distance ΔQ separating the AP minima for the PJT center in the e - and h -state that results in a considerable positive isotope coefficient (IC) at low T_c . In a multi-mode case the g factor contribution plays the same role, but the D factor contribution can be both positive and negative. The principal results of analytical consideration and computer simulation for the vibronic IE on the local boson transfer integral and T_c in the system of the PJT centers could be summarized as follows:

1. There is no simple $\alpha(T_c)$ correlation, nonetheless the absolute value of the IC usually increases with decreasing T_c . Minimal IE should be observed for the optimized systems with maximal T_c 's. However, this doesn't imply a negligible role of the electron-vibrational coupling. Within our model an

extremely strong electron-vibrational (vibronic) coupling easily coexists with the negligibly small IE.

2. As a whole, the vibronic contribution to IE appears to be relatively small, so even at favorable conditions the oxygen exponent α_O reaches the corresponding BCS value $\alpha_{BCS} = 0.5$ only for small $T_c \sim 0.2 T_c^{max}$. Anomalously large values $\alpha_O > 1.0$ are possible only at $T_c < 0.1 T_c^{max}$.

3. As a rule, both oxygen and copper IC appear to be positive. Negative IE could be observed as a result of peculiar compensation of the positive g - and negative D -contributions. In practice, such a compensation could be realized only for copper due to occurrence of the only hybrid e_u modes and within a rather narrow range of the appropriate parameters and with a rather small numerical value $|\alpha_{Cu}| \leq 0.2$. Such a compensation is unlikely simultaneously for several oxygen modes. Quite the contrary, negative copper IE $\alpha_{Cu} \leq 0$ is usually accompanied by relatively large positive α_O values, while positive copper IE $\alpha_{Cu} \geq 0$ does by moderate positive α_O values.

4. Numerical values of the copper and oxygen IC's are determined by the material-dependent quantities such as bare parameters Δ_e, Δ_h for the $^1A_{1g}-^{1,3}E_u$ separation, elastic and vibronic constants. Moreover, for instance, the most popular HTSC systems $YBa_2Cu_3O_{6+x}$ and $La_{2-x}Sr_xCuO_4$ are distinguished by the ground state adiabatic potential ($b_{1g}e_u$ - and $b_{2g}e_u$ type, respectively) that easily explains an observed difference in numerical values and, even, in sign relations for the corresponding copper and oxygen IC's^{4,5}.

For accounting of the pressure effect on T_c through the K_{eh} one need to determine full variation of the ground vibronic state of the PJT center including the pressure induced electronic density redistribution. The pressure effect for the polar CuO_4 -center can be described by an effective operator

$$\hat{U}_p = \sum_i p_i^{(m)}(\mathbf{p}) \hat{Q}_i + \sum_i p_i^{(e)}(\mathbf{p}) \hat{V}_i, \quad (7)$$

where the first term describes the purely mechanical perturbation, and the second one corresponds to a pressure induced low-symmetry crystalline field, Q_i and \hat{V}_i are the distortion modes and electron operators with a certain symmetry, respectively. To get a linear in pressure variations $\delta\varepsilon_0$ and $\delta\varphi_0$ to the vibronic ground state energy ε_0 and wave function φ_0 in a framework of the Opik-Price method, one needs to solve the eigenvalue problem for the potential energy matrix

$$\hat{U}(Q) = \hat{U}_0(Q) + \delta\hat{U}_p, \quad (8)$$

$$\hat{U}_0(Q) = \hat{H}_{el} + \sum_i \frac{\omega_i^2 Q_i^2}{2} \hat{I} + \sum_i \hat{V}_i Q_i, \quad (9)$$

$$\delta\hat{U}_p = \sum_i \delta p_i^{(m)} Q_i \hat{I} + \sum_i \delta p_i^{(e)} \hat{V}_i, \quad (10)$$

in the AP minima with taking account of the linear in pressure variation of their coordinates:

$$Q_i = Q_i^{(0)} + \delta Q_i, \quad \delta Q_i = -\frac{\delta p_i^{(n)}}{\omega_i^2} - \frac{2\langle\varphi_0|\hat{V}_i|\delta\varphi_0\rangle}{\omega_i^2}. \quad (11)$$

The system for $\delta\varepsilon_0$ and $\delta\varphi_0$ takes a form

$$\begin{aligned} (\hat{H}_{el} + \sum_i \hat{V}_i Q_i^{(0)})|\delta\varphi_0\rangle + \sum_i \hat{V}_i (\delta Q_i + \delta p_i^{(e)})|\varphi_0\rangle &= \lambda_0|\delta\varphi_0\rangle + \delta\lambda_0|\varphi_0\rangle \\ \langle\varphi_0|\delta\varphi_0\rangle &= 0, \quad \delta\varepsilon_0 = \delta\lambda_0 - 2\sum_i \langle\varphi_0|\hat{V}_i|\delta\varphi_0\rangle Q_i^{(0)}. \end{aligned} \quad (12)$$

In addition, pressure results in a variation of the energy surface form near a certain minimum of the adiabatic potential

$$\begin{aligned} \delta\epsilon &= \sum_i \frac{2\langle\varphi_0|\hat{U}_1|\varphi_i\rangle}{\lambda_0 - \lambda_i} \left[\langle\delta\varphi_0|\hat{U}_1|\varphi_i\rangle + \langle\varphi_0|\hat{U}_1|\delta\varphi_i\rangle \right] \\ &\quad - \sum_i \frac{\langle\varphi_0|\hat{U}_1|\varphi_i\rangle^2}{(\lambda_0 - \lambda_i)^2} (\delta\lambda_0 - \delta\lambda_i), \end{aligned} \quad (13)$$

where \hat{U}_1 is the first variation of the vibronic Hamiltonian relative to Q_i ; φ_i , λ_i and $\delta\varphi_i$, $\delta\lambda_i$ are the eigenvectors and eigenvalues of the non-perturbed Opik-Price system and their variations under pressure, respectively. The system for $\delta\varphi_i$, $\delta\lambda_i$ is quite similar to (12). The second term in (13) results in a renormalization of the local frequencies, while the first one provides a hybridization of all the local modes. The relations (11)-(13) together with (3) provide an algorithm for a calculation of the pressure dependence of the vibronic reduction factors.

In framework of the developed approach we have performed a number of model calculations of the baric coefficients with reasonable values of various parameters. Their results will be published elsewhere. As a whole, the PJT nature of the system explains both the main peculiarities and subtleties of the pressure effects including a strong anisotropy of the baric coefficients in 123 system.

At once, a real situation with isotope and baric effects in doped cuprates should account for phase separation and percolation phenomena.

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